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Part 1: Impact on Pollutant Formation

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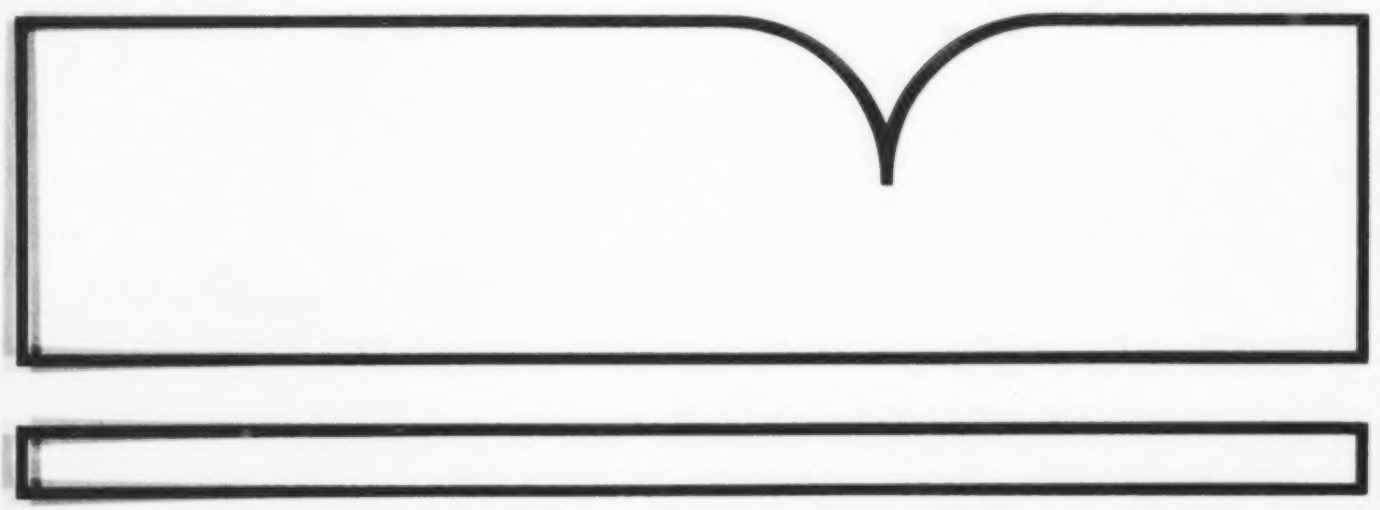

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16. ABSTRACT The paper gives results of a study of the ignition process of liquid fuel droplets and its impact on the formation of soot and nitrogen oxides (NOx). The system studied consists of a laboratory flame maintained by a vertical, monosized liquid fuel droplet array surrounded by a laminar flow of a mixture of helium and oxygen. It was found that the formation of soot and NOx closely correlated with the ignition of liquid fuel droplets. It has been shown that the soot formation can be suppressed by delaying ignition, and thus vaporizing as much fuel as possible prior to the ignition. But delaying ignition favors the conversion of fuel-bound nitrogen to nitric oxide (NO). The control of the emission of both soot and NOx can be achieved by use of low oxygen concentrations in the oxidation gas to delay ignition and suppress soot formation by using a high fuel/air ratio to reduce NOx.

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Combustion	Nitrogen Oxides	n-Dodecane	
Quinolines			07C
Helium			07B
Oxygen			

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Ignition and Combustion of Liquid Fuel Droplets. Part I: Impact on Pollutant Formation

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Abstract—The ignition process of liquid fuel droplets and its impact on the formation of soot and nitrogen oxides have been studied. The system studied consists of a laboratory flame maintained by a vertical, monosized liquid fuel droplet array surrounded by a laminar flow of a mixture of helium and oxygen. It was found that the formation of soot and nitrogen oxides closely correlates with the ignition of liquid fuel droplets. It has been shown that the soot formation can be suppressed by delaying ignition, and thus vaporizing as much fuel as possible prior to the ignition. But delaying ignition favors the conversion of fuel-bound nitrogen to nitric oxide. The control of the emission of both soot and nitrogen oxides can be achieved by use of low oxygen concentrations in the oxidation gas to delay ignition and suppress soot formation by using a high fuel/air ratio to reduce NO_x.

INTRODUCTION

Liquid fuels are injected into combustion chambers through atomizers to facilitate the disintegration of the liquid into a spray of droplets. When the fuel vapor is ignited in the vicinity of the droplets this results in an increased rate of evaporation. Reaction zones will be formed partly surrounding each droplet or in a region where fuel/air ratios are within limits of inflammability. To obtain a full description of the processes involved in spray combustion it is necessary to have complete knowledge of the mechanism of combustion of the individual droplets, any interaction between the droplets, and the statistical description of the droplets with regard to size and spatial distribution. It is difficult to obtain detailed information on the mechanism or rate of burning by direct studies of spray combustion. A number of experimental techniques have been developed to study the combustion of isolated single droplets under carefully controlled conditions. Theoretical analyses and mathematical models are also greatly assisted if only single isolated droplets are considered (Law, 1982). However, the single droplet experiment, especially with a single droplet suspended at the end of a thin quartz fiber, has several limitations. Free droplet array experiments offer the advantages of small size droplet, noninterference from suspension fiber, and the capability of using volatile fuels. Also, the regularity of the droplet stream produced permits detailed photographic observations using stroboscopy.

The effect of interaction between droplets in the droplet array can be analyzed theoretically (Labowsky and Sangiovanni, 1980) by making correction on the evaporation rate constant obtained for the single droplet. The correction for the external convection effect can also be made (Frössling, 1938; Ranz and Marshall, 1952).

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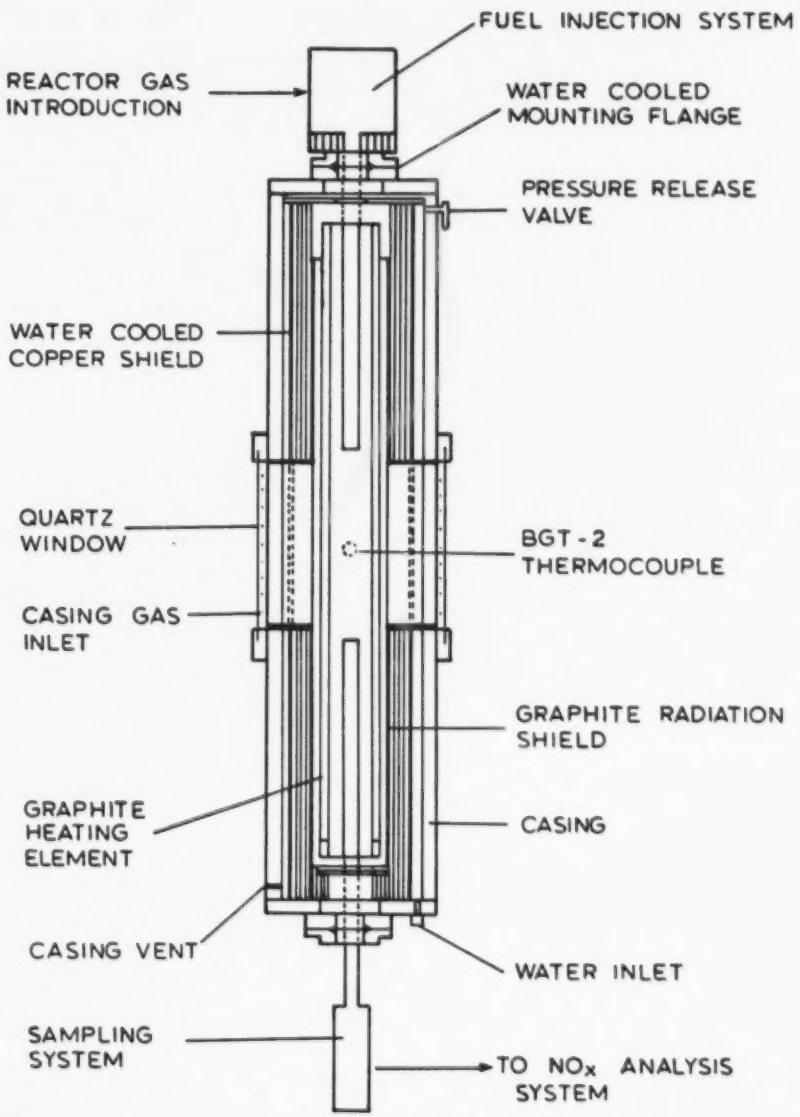

FIGURE 1 Schematic of experimental system.

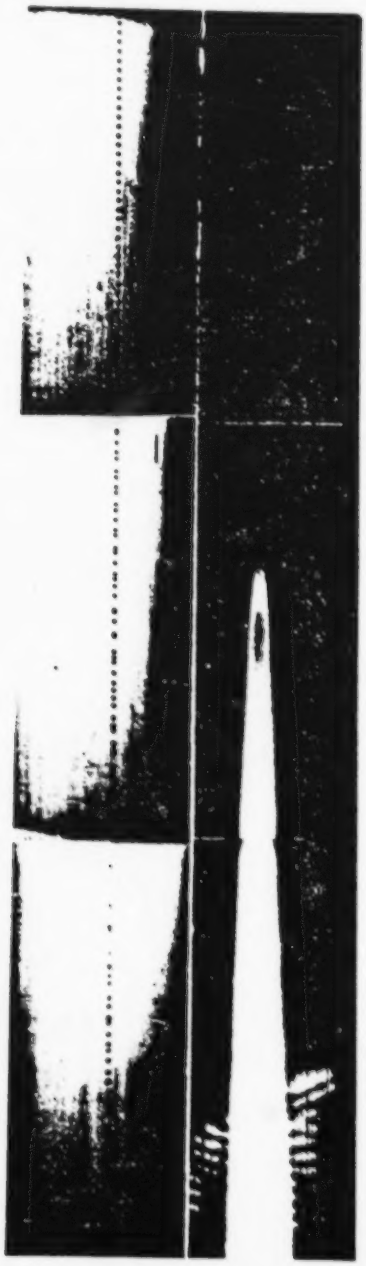

FIGURE 2 Photographs of a burning droplet array.

Combustion products appearing only in trace concentrations include nitric oxide (NO), nitrogen dioxide (NO_2), sulfur dioxide (SO_2), and unburnt or partially reacted hydrocarbons including polycyclic aromatics, soot and metallic compounds. The present study is restricted to nitrogen oxides and soot.

By far the most attractive NO_x control strategy involves modification of the combustion process by employing a staged combustion technique (Beér *et al.*, 1983). In this technique the major proportion of the fuel-bound nitrogen is converted in a fuel-rich zone to relatively inactive molecular nitrogen. Complete combustion of the fuel is accomplished in a subsequent fuel-lean zone in which carefully controlled conditions minimize the formation of thermal NO. Although the staged combustion technique is the most promising strategy for NO_x control, there is concern over soot emission from the staged combustion of fuels of highly aromatic and high C/H ratio contents. The operating conditions in the fuel-rich first stage may result in the formation of a significant amount of soot. There is a definite trade-off between NO_x control and soot formation in the staged combustion (Blazowski *et al.*, 1981).

The formation of nitrogen oxides and soot in the fuel-rich first stage of a staged combustor provides a motivation of this study. In preliminary studies, it has been found that the ignition of liquid fuel droplets and the formation of an envelope

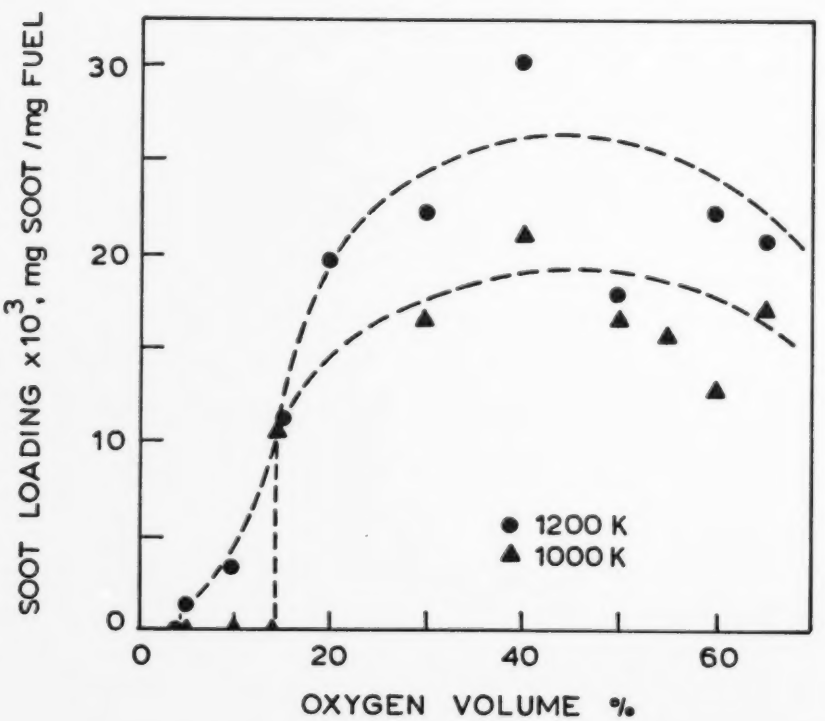

FIGURE 3 Soot loading vs. oxygen concentration.

flame around the droplet array have profound effects on the formation of nitrogen oxides and soot. The objective of this study is to understand the ignition process of liquid fuel droplets, to see how this ignition process influences the formation of soot and nitrogen oxides from oil droplet flame, and by a theoretical modeling, to confirm experimental observations on the ignition process.

EXPERIMENTAL PROCEDURE

The experimental apparatus selected is illustrated in Figure 1 (Hanson, 1982; Rah, 1984). The major components of the system are: a laminar flow furnace; a fuel reservoir and atomizer; a sampling system; a gas flow metering and control system; a photographic device; an optical system for soot concentration measurement; a soot recovery (by filtration) and weighing system; and a NO/NO_x analyzer system.

A monosize linear array of liquid fuel droplets was introduced into the isothermal furnace (Astro) by means of a vibrating orifice atomizer. The droplet array was surrounded by a laminar flow of a mixture of helium and oxygen of specified concentration. The residue of fuel droplets was collected through a transpiring wall sampling probe equipped with a cascade impactor (Pilat Mark 3). A gravimetric method and an optical transmittance measurement technique were employed to determine soot loadings for various experimental conditions. Nitrogen oxides emissions were

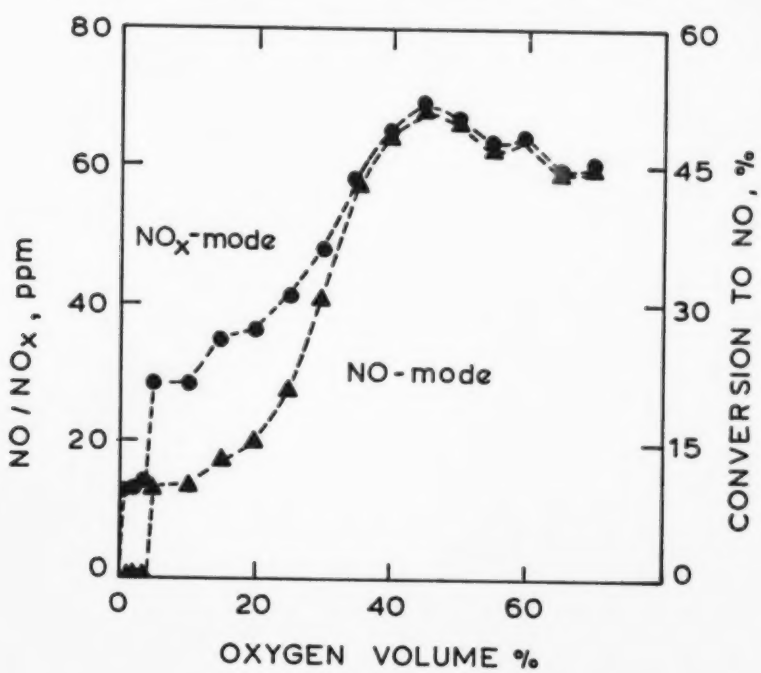

FIGURE 4 NO emission vs. oxygen concentration.

measured from burning droplets by using a chemiluminescent NO–NO_x analyzer (Thermo Electron Model 14A). A record of droplet size and ignition distance, which could be translated into ignition delay time, was obtained photographically.

An optical system using light absorption technique was developed and employed to measure the soot concentration in the flame surrounding droplet arrays. The system uses a collimated beam of width greater than that of the flame for ease of alignment. A generalized treatment of the problem involving transmittance of collimated radiation by objects of various shapes is reported by Rah *et al.* (1984).

Depending upon oxygen concentration and furnace temperature, the luminosity of the flame changed drastically. In Figure 2, photographs of a burning droplet array of n-dodecane taken by using two different techniques are presented. The picture on the left-hand side taken by using a stroboscope synchronized with the droplet generator shows the behavior of individual droplets. We can observe the flame shape and the ignition distance from the picture on the right-hand side. When the droplets are initially away from the ignition point, the spacing between the droplets are uniform. However, the spacing becomes uneven as the droplets approach the ignition point due to severe interactions between droplets. Because of the aperture limitations, only the upper portion of the flame can be displayed.

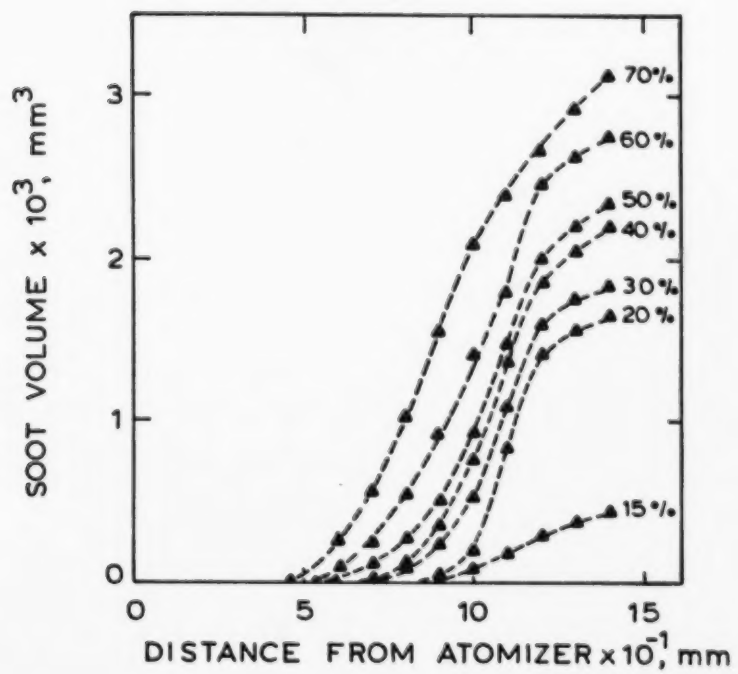

FIGURE 5 Soot volume profile for various oxygen concentrations (1000 K).

RESULTS AND DISCUSSION

Relation of Soot and NO_x Emission to Ignition

The amount of soot recovered (mg) from the filtration process per unit mass (mg) of fuel (n-dodecane) injected for two different furnace temperatures with respect to various oxygen concentration (in volume percent of oxygen in the mixture of helium and oxygen) is plotted in Figure 3.

For both temperatures there are certain limits of oxygen concentration over which stable visible flames were obtained. At oxygen concentrations below the limit at which no visible flame could be observed no soot was collected. This clearly shows the importance of ignition phenomena in relation to the formation of soot in liquid fuel combustion process.

As we increase the oxygen concentration, the amount of soot collected increases and then decreases with a maximum of soot loadings around 40 percent of oxygen for both furnace temperatures. This may be interpreted as follows: the flame temperature increases with increased oxygen concentration, resulting in the higher rate of pyrolysis of soot precursors and thus the greater tendency to soot; at the same time the rate of soot oxidation increases with increases in oxygen concentration (Glassman and Yaccarino, 1981).

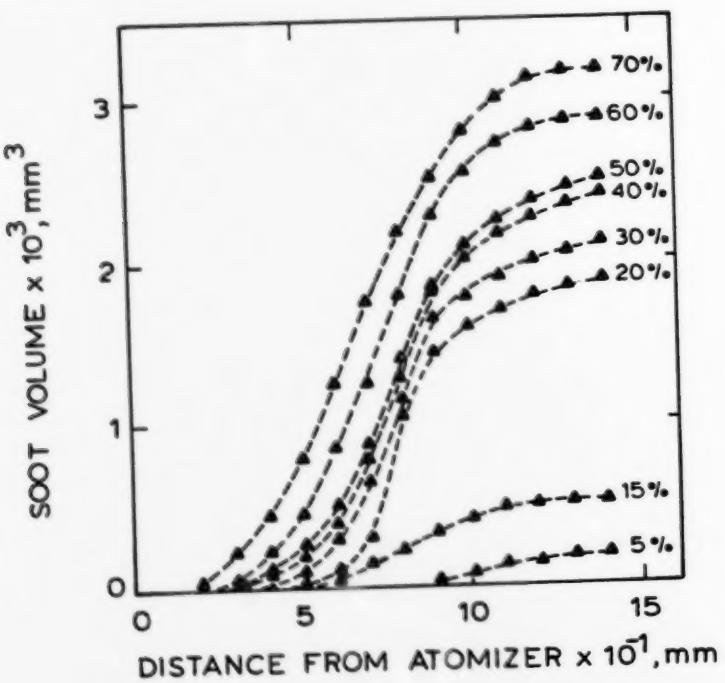

FIGURE 6 Soot volume profile for various oxygen concentrations (1200 K).

The fuel used for the NO_x emission experiment was n-dodecane doped with quinoline. To simulate the nitrogen content of typical high nitrogen-containing fuels the weight percent of nitrogen was chosen to be 1. No difference in the soot loading between this doped fuel and the undoped fuel was observed. In Figure 4 the NO/NO_x concentrations in the combustion products of 100 μm droplet arrays are presented for various oxygen concentrations. Also shown are the corresponding conversions of fuel nitrogen to NO. The maximum conversion is about 55 percent, which is of similar level to data from other studies on diffusion flames. A minimum of 5 percent oxygen in the gas stream is necessary for ignition. In the pre-ignition zone fuel evaporates and the vapor is mixed with oxygen. The ignited droplet array then burns in the form of an envelope flame. The difference between the NO_x-mode

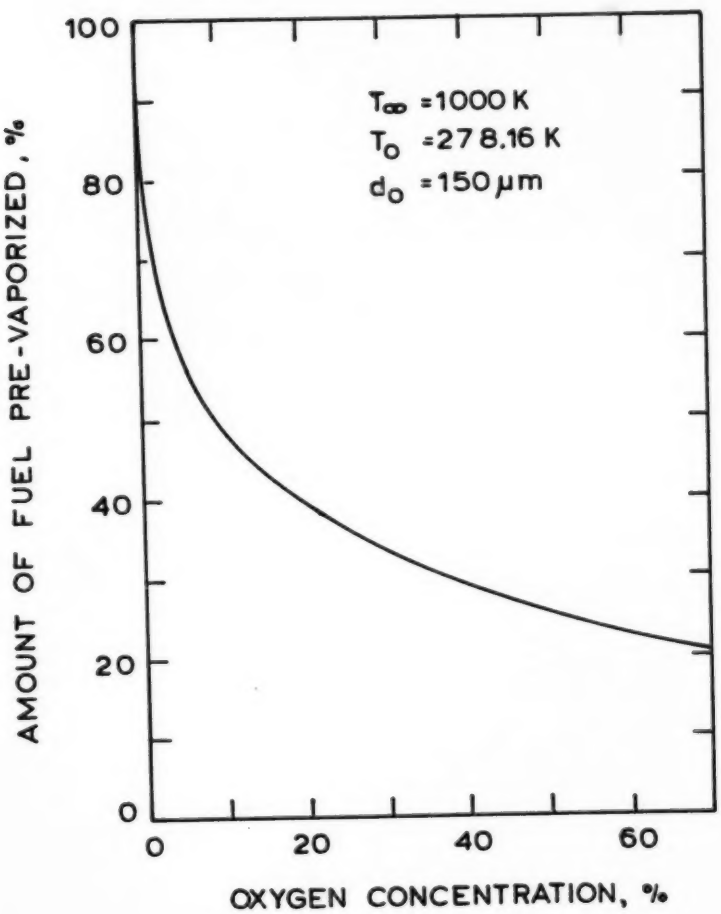

FIGURE 7 Amount of fuel vaporized before being ignited vs. oxygen concentration.

reading and NO-mode reading in Figure 4 represents concentrations of the nitrogenous intermediates converted in the chemiluminescent NO–NO_x analyzer with stainless steel catalyst (Hodgeson *et al.*, 1971). The sudden jump in the reading at the igniton point is caused by a surge in the evaporation rate of fuel.

Ignition Studies

We have observed that the emission of soot and NO_x closely correlate with the ignition of the fuel droplets. In the present study, an attempt is made to analyze the problem of liquid fuel droplet ignition with the reasoning based on the thermal theory of ignition of gaseous fuels (Merzhanov and Averson, 1971; Gray and Sherrington, 1977). By specifying the droplet surface position with the d^2-law, a system of partial differential equations describing the ignition process could be solved numerically. The effects of convection and interference between droplets in a linear array of droplets were lumped on the evaporation rate constant in the d^2-law. This method of approach, from an analytical point of view, has the advantage of using the spherical

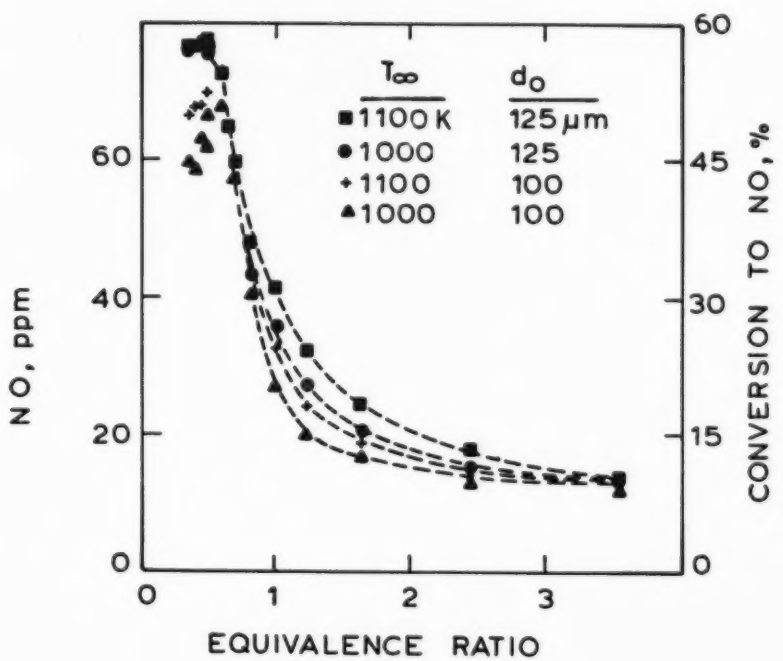

FIGURE 8 Effect of furnace temperature and initial droplet size on NO emission.

symmetry of the system. The model served for the interpretation of the experimental results on the ignition of pure fuel, and by extension, of heavy fuel oils. Details of the ignition studies are presented in a companion paper (Rah *et al.*, 1985).

Practical Implications

The ability of the ignition model to predict the effect of oxygen concentration on the point of ignition in experiments on soot formation is shown in Figures 5 and 6. Each 10 mm segment of the flame along the droplet array axis was optically observed. The data were extrapolated to the ignition point determined from calculations, where the soot concentration was considered to be zero. The general trend in the changes of soot volume along the droplet array axis is similar for both temperatures: a change in the furnace temperature causes a change in the ignition pattern of the fuel. However, the sooting character of the system does not change drastically, because the energy for the pyrolysis is mainly provided by the stationary flame formed around the droplet array, and is little influenced by heat transfer from furnace wall. This shows that the flame temperature is a significant factor which determines the propensity to soot in the droplet burning.

The amount of soot formed decreases as we decrease the oxygen concentration. In practical combustors, this can be achieved by recirculation of flue gas. This can be explained by the increase in the fraction of fuel vaporized prior to ignition with decrease in the oxygen concentration. From the ignition model discussed in the companion paper we can estimate the amount of fuel evaporated prior to ignition (Figure 7). At 5 percent oxygen concentration, for example, more than 55 percent of the fuel is predicted to vaporize before ignition. However, this increase in prevaporization and premixing before ignition poses a potential problem for NO_x formation. But, by keeping fuel/air ratio high as well as the oxygen concentration low we can take advantage of both lowering soot and NO_x formation.

Presented in Figure 8 are the effects of overall equivalence ratio on conversion of fuel nitrogen to NO for different droplet sizes and furnace temperatures. We can observe a decrease in the conversion efficiency with an increase in the equivalence ratio. As long as we have an envelope flame surrounding the droplet array, it is the overall equivalence ratio that determines the NO emission from fuel nitrogen. This effect of the equivalence ratio has been recognized from studies on premixed flames (Fenimore, 1972; Sarofim *et al.*, 1975), on oil-doped spray combustors (Turner *et al.*, 1972; Hazard, 1973; Flagan and Appleton, 1974), and on a coal combustor (Pohl and Sarofim, 1977). The slight increase in NO emission at higher furnace temperature is a composite effect of the increased rate of evaporation and the subsequent gas phase oxidation.

There are two degrees of freedom for the designer: (a) the oxygen concentration controllable by flue gas recycle, and (b) fuel/air ratio. Theory can provide guidance to these selections.

CONCLUSIONS

The formation of soot and nitrogen oxides closely correlates with the ignition of fuel droplets. Substantial yields of soot are produced when an envelope flame is formed around the burning droplets, increasing dramatically as the oxygen concentration is increased beyond that required for ignition. Our observations show that the flame temperature is also an important parameter for the tendency to soot.

For the NO emission, the overall equivalence ratio is the important factor as long as there is an envelope flame around the droplets.

Soot formation can be suppressed by delaying the ignition and the formation of an envelope flame around the droplets, vaporizing thereby as much fuel as possible before ignition. Ignition delay, however, may cause more NO_x to form. By keeping the fuel/air ratio high for reduced NO_x emission and using recirculation of oxygen-depleted combustion products for suppressing soot formation means can be found to control the emission of both these pollutants.

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